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AN ANALYSIS OF THREE PULTRUSION MODELS PROPOSED FOR USE IN A PULTRUSION CONTROL SYSTEM

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COMPOSITES DEVELOPMENT BRANCH

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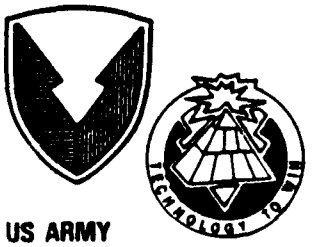

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ABSTRACT

Three mathematical models currently under consideration as candidates for use in real-time control of the pultrusion process have been implemented and compared. Simulations of the pultrusion process were performed using lumped parameter, one-dimensional finite difference, and two-dimensional finite difference models. The results of the simulations were compared to each other, and to an experimentally acquired thermal history of pultruded glass/epoxy bar stock, in order to determine the suitability of the various models to the process and control environment. The die temperature profile used in the models was experimentally determined as was the initial composite temperature. Kinetic parameters for the anhydride hardened, amine accelerated epoxy Diglycidyl Ether of Bisphenol A (DGEBA) resin system were obtained using data from a previously published study which employed differential scanning calorimeter (DSC) analysis.

The sensitivity of the models to variations in material properties and kinetic parameters was also investigated. The models were run with values of thermal diffusivity, heat of reaction, rate of reaction, and activation energy which were varied by amounts within the realm of experimental error. The results are reported in terms of maximum exotherm temperature and the position within the die at which this occurs.

A comparison of the three models was made in terms of run time on two PC based platforms. A 6 MHz 80286AT class computer equipped with an 80287 math coprocessor, and a 16 MHz 80386SX computer equipped with the appropriate math coprocessor, were used for these studies.

Finally, a methodology for the inclusion of a mathematical model in a real-time robust control system is discussed.

NOMENCLATURE

α	=	degree of cure
$\bar{\alpha}$	=	lumped degree of cure
ρ	=	density
A	=	pre-exponential
C_P	=	heat capacity
E	=	activation energy
Hr	=	heat of reaction
k	=	thermal conductivity
l_x	=	height of stock
ly	=	width of stock
R	=	gas constant
T	=	temperature
$\bar{T}$	=	lumped temperature
t	=	time
X	=	distance in x coordinate
Y	=	distance in y coordinate

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INTRODUCTION

Pultrusion Process Control

As one of the few continuous composites processing techniques available, pultrusion offers the advantage of a less expensive method of fabrication than alternative processes. However, the continuous nature of pultrusion, while offering cost efficiency inherent in continuous processes in general, presents problems in process control not encountered in continuous fully liquid processes. In liquid processes, temperatures, pressures, and various other parameters can be directly measured during processing and used in real-time process monitoring and control without affecting the quality of the product. The plug flow nature of the pultrusion process, however, is not conducive to continuous process monitoring since internal sensors which, when used, necessarily become incorporated into the product.

Embedment of sensors is problematic. If left in a final part they may be detrimental to the material properties. The alternative is the disposal of those portions of product containing sensors. Recently, a great deal of effort has been extended toward developing compatible and nondestructive sensors for inclusion in composite structures. These may be dual purpose sensors intended to serve both as an aid during processing and as monitoring devices during the structure's service lifetime. While this approach has merit, the successful development of compatible sensors does not eliminate all problems of embedded sensors.

Experience with embedded thermocouples has shown that accurate placement of embedded sensors can be troublesome. Although this technique can be useful for estimating the maximum temperature achieved within the pultruded stock, it is limited since the location of the sensor cannot be accurately determined as it is placed into the fiber/resin mass. Reliable sensor placement also becomes increasingly difficult as line speeds increase. Although this procedure can be useful for some purposes, the time required to insert the sensor, allow it to pass through the die and pullers and subsequently determine its location, prevents it from being a useful tool in real-time process control.

One possible solution to the problem of determining conditions within the composite stock as it passes through the die is to use a process model. It is the purpose of this work to investigate the feasibility of using this approach to generate internal temperature profiles for use in real-time process control.

In order for this method to be considered feasible, the following criteria must be met:

- The material properties, kinetic properties, etc., used in the model must be readily available or easily determined.
- The model results must be reasonably accurate
- The time required to execute the model on the chosen computer platform must be reasonable in terms of feedback control.
- The computing machinery must be relatively inexpensive.
- Mechanisms must be in place to ensure that solutions converge and that program exceptions are handled in a rational manner.

The efforts detailed in this report encompass the development and execution of three models, production of experimental data, and an evaluation of the three models based upon the criteria outlined above.

Materials and Processing Equipment

The system formulation selected for this study consisted of 100 parts Epon 826 resin, a Diglycidyl Ether of Bisphenol A (DGEBA, manufactured by Shell Chemical Company), 80 parts Methyl Tetrahydro Phthalic Anhydride (MTHPA, manufactured by Anhydrides and Chemicals, Inc.) as the hardener and one part Benzyldimethylamine (BDMA, manufactured by Miller Stephenson Chemical Company, Inc.) as the accelerator, in conjunction with an internal mold release (CEARA HT-1, made by Ceara Products, Inc.) at 1.5% of total weight and a viscosity modifier Zeothix 265 (a precipitated amorphous silica, produced by J. M. Huber Corporation) at 2.5% of the total weight. Both E- and S-glass fiber rovings treated with an epoxy compatible sizing were used to produce pultruded stock having calculated theoretical fiber volume of 65%.

The pultrusion equipment used at the U. S. Army Materials Technology Laboratory (MTL) is a Goldsworthy Glastruder. The rovings are dispensed from a creel, which contains the tows either in large center wrapped packages or on individual spools; the fibers then pass through a carding rack and into a resin bath. Upon leaving the bath the fibers pass through a set of Teflon® rollers designed to squeeze out excess resin and into a series of three Teflon® preformers. The preformers, spaced about two feet apart, aid in resin wet-out of the fiber mass. Having achieved the desired wet-out and approximate shape, the fiber bundle enters the heated die. The die used in this study is 30" in length and produces rectangular stock of a 1/4" x 1" cross-sectional area. Die heating is accomplished by means of induction heaters located in three zones along the die length. The heaters are controlled through thermocouples imbedded in the die at approximately the center position of each temperature zone. In situ temperature data was acquired using an industrially hardened IBM PC-AT computer in conjunction with a Cyborg Loggernaut® data logger.

Thermocouples were implanted within and on the surface of the fiber mass just prior to its entry into the die. The exact position of these sensors was determined when the specimens were cut during post processing analysis.

MODELS

Description of the Mathematical Models Used

The solution to the mathematical description of the temperature and conversion profiles for the pultrusion process has been arrived at through the implementation of a lumped parameter, a one-dimensional and a two-dimensional model. The three models are essentially the same with the exception of dimensionality. Each makes the same set of simplifying assumptions. The effects of back resin flow and axial heat conduction are assumed to be negligible. Thermal properties and constants used in describing reaction kinetics are assumed to remain constant throughout the process. It is also assumed that both fiber and resin within the same locality have the same value of temperature and that the pultruded stock is in good thermal contact with the die. The governing equations are:

$$\rho C \frac{dT}{dt} = \nabla \cdot (k \nabla T) + \rho \ Hr \frac{d\alpha}{dt} \quad (1)$$

$$\frac{d\alpha}{dt} = (1-\alpha) \ A \exp(-E/RT) \quad (2)$$

At $t=0$, $\quad 0 \geq X \geq 1x$, $\quad \alpha = 0$

At $t \geq 0$, $\quad X = 1x$, $\quad T = T_D$

where,

ρ	=	the density (gm/cc)
C	=	the heat capacity (cal/gm K)
k	=	the thermal conductivity (cal/cm K sec)
∇	=	the del operator
Hr	=	the heat of reaction (cal/cm^3)
α	=	the degree of cure
A	=	the pre-exponential
E	=	activation energy (cal/mole)
R	=	gas constant (cal/mole K)
T	=	temperature K
lx	=	height of stock
Td	=	the die temperature

The lumped parameter model[1] implements the solution to the following equations:

$$\rho \ Cp \frac{d\bar{T}}{dt} = \frac{3k}{\frac{1}{2}x} (T_D - \bar{T}) + \rho \ Hr \frac{d\bar{\alpha}}{dt} \quad (3)$$

$$\frac{d\bar{\alpha}}{dt} = (1 - \alpha) \ A \exp(-E/R\bar{T}) \quad (4)$$

where,

$\bar{T}$	=	lumped or average slab temperature
$\bar{\alpha}$	=	lumped degree of cure

Equations 3 and 4 are derived from Equations 1 and 2 by assuming a parabolic temperature distribution in the thickness direction. The solution employs a time-stepping scheme in which the differential Equations 3 and 4 were solved using the Runge-Kutta Algorithm. Although solution of the lumped parameter model yields an average slab temperature, the assumption of a parabolic profile allows for the calculation of a temperature and conversion profile throughout the thickness of the pultruded stock.

1. OSWTON, S. G. *Pultrusion Process Thermal Modeling.* TASC IOM-SGO-86-01, 1986.

The one-dimensional and two-dimensional models employ a fully implicit control volume finite difference solution based upon methods developed by Patankar.[2] They were written specifically for the pultrusion process in order to eliminate unnecessary calculations which might be performed by a more general model. Each takes advantage of the Thomas or Tri-Diagonal-Matrix Algorithm (TDMA) for the solution of a source linearized version of Equation 1 in a single dimension. In the one-dimensional case Equations 1 and 2 are resolved through an iterative scheme. The two-dimensional solution requires iteration to resolve, not only Equations 1 and 2, but also to resolve TDMA solutions in the x and y directions. As in the case of the lumped parameter model, the solution is time stepped.

In each model the time step is constant and nodes in the x direction are equidistant as are the nodes in the y direction. Included in the one-dimensional model is an automatically adjusted relaxation term which is activated when the difference between the current estimate of temperature and the previous estimate is too great. This condition might otherwise lead to an overflow exception which would cause the program to abort. The relaxation term slows the iteration by reducing the difference between the current and previous temperatures and using an intermediate temperature as an estimate, thus insuring that excessive overshoot does not occur. The solution for the one-dimensional model is given in the Appendix.

The language chosen was Borland's Turbo Pascal.® This language was chosen for compatibility reasons (future incorporation into an existing control program written in Turbo Pascal® is envisioned).

Model Verification

Each of the models written for this study was verified through comparisons to existing models which are known to be correct. Results of the lumped parameter model were compared to solutions arrived at through the use of the HEATING5 computer code.[3] A comparison of the temperature history and conversion fractions revealed that the maximum discrepancy between the two methods is about 20°C occurring at early times. Overall, there is good agreement with only a 5°C difference near the peak exotherm. The solution for the one-dimensional model with no heat source was validated by comparison to the classic heat transfer solution for conduction in a semi-infinite solid as described in Bird, Stewart, and Lightfoot.[4] The temperature profile was the same for both the one-dimensional and the classical solution. Results from the two-dimensional model were compared to the solution from a previously implemented two-dimensional model.[5] The two solutions were practically identical.

THERMAL AND KINETIC PARAMETER DETERMINATION

Determination of Physical Properties

One of the simplifying assumptions made in formulating the models was that thermal and kinetic properties do not change with temperature or degree of conversion. In addition to

2. PATANKAR, S. V. *Numerical Heat Transfer and Fluid Flow*. McGraw-Hill Book Co., New York, NY, 1980.
3. TURNER, W. D., ELROD, D. C., and SIMON-TOV, I. I. *HEATING5 - An IBM 360 Heat Conduction Program* Oak Ridge National Laboratory, ORNL/CSD/TM-15, 1977.
4. BIRD, R. B., STEWART, W. E., and LIGHTFOOT, E. N. *Transport Phenomena*. John Wiley and Sons, Inc., New York, NY, 1960, p. 354-357.
5. WALSH, S. M., and CHARMCHI, M. *Heat Characteristics of a Pultrusion Process*. 25th National Heat Transfer Conference, Houston, TX, 1988.

limiting the complexity and execution time of the models, this assumption ensures that the time and effort required to determine these properties is not excessive. The properties used in the models were determined from samples of the cured stock which were produced under the same boundary conditions as those used in the running models. The line speed was six inches per minute and a control temperature of 330°F was used for all three die zones. The specific properties of interest were the density, specific heat, and thermal diffusivity; the product of which yields thermal conductivity, as shown in Equation 5.

$$K = \alpha \rho C_P \tag{5}$$

The fiber volume fraction was also of interest and was determined by resin burnout tests which were performed on finished pultruded stock.

The thermal diffusivity of the material was determined by the flash pulse method. The method was developed by Parker, et al. in 1961[6] and allows one to directly determine the thermal diffusivity of a material. Briefly, the experimental procedure involves exposing the front face of a thin sample to a short pulse of radiant energy. The temperature history of the back face is recorded, and from it the thermal diffusivity can be determined. For this study, the determination was made using the half-time method which calculates the diffusivity from the time required to reach one-half of the maximum back surface temperature. The governing relationship is given by:

$$\alpha = \frac{1.38L^2}{\pi^2 t_{0.5}} \tag{6}$$

where,

α = the thermal diffusivity
L = the sample thickness
$t_{0.5}$ = the time to reach half the maximum back surface temperature

Several conditions must be met for this technique to be viable. There can be no phase changes in the sample and the thermal diffusivity of the sample should remain constant over the temperature rise experienced by the sample during testing. Also, the heat pulse must be of short duration compared with its time of passage through the sample and this passage must be sufficiently short so that heat losses will be negligible. The method has been applied to composite materials[7,8] where an effective value for a specific fiber/resin content is determined.

One of the advantages of the method is that it translates rather easily into an experimental apparatus. The equipment used for determining diffusivity values in this study consisted of an Ascorlight, Model No. QC8, xenon flash tube with a power source, a spring loaded open junctioned chromel-alumel thermocouple equipped with an electronic ice point, a

6. JENKINS, R. J., and PARKER, W. J. *Flash Method of Determining Thermal Diffusivity, Heat Capacity and Thermal Conductivity.* WADD Technical Report No. 61-95, U. S. N. Rad. Def. Lab., Project No. 7360, June 1961.
7. TAYLOR, R. E., JORTNER, J., and GROOT, H. *Thermal Diffusivity of Fiber Reinforced Composites Using the Flash Technique.* Carbon, v. 23, no. 2, 1985.
8. JAKLITSCH, D. J., and WALKINSHAW, J. W. *Flash Pulse Measurement for Off-Axis Thermal Conductivity of Carbon Composite Materials.* Ind. Eng. Chem. Res., v. 27, no. 4, 1988.

5000 series Tektronics digital oscilloscope containing a differential amplifier, and a Hewlett Packard X-Y recorder which was used to make a permanent record of the oscilloscope trace. The samples were coated on the front face with a thin layer of a carbon black based mixture (50 parts carbon black, 25 parts carboxymethylcellulose, and 25 parts tetrasodiumpyrophosphate) in order to insure good and uniform heat absorption. In order to insure good electrical contact between the thermocouple and the back face of the composite sample, an additional conductive layer was added using an electrically conductive silver paint. The possible effects of coatings in general are described in Reference 8. In this case the coatings were thin enough, or like the silver paint, more conductive than the sample so that they had no significant effect on the measurement of diffusivity.

A series of eight samples of various thicknesses were used in the experiments in which a total of 27 values of thermal diffusivity were determined.

The heat capacity of the composite was determined using a Perkin Elmer Differential Scanning Calorimeter (DSC), Model DSC-2. The method used involves the comparison of two samples of similar weight, a known sample (sapphire in this case) against the unknown sample. The temperature range of the scan was approximately 20 K with the desired temperature of interest falling about midway in the scan. The procedure uses a comparison of the amplitudes of the curves which are determined from the differential heat energy input into the pans. Four samples were examined with an average weight of about 32 milligrams ranging from a low of 25.9 milligrams to a high of 37.4 milligrams. The sapphire reference weighed 35.4 milligrams. Three separate scans were carried out on each of the samples, as well as on the sapphire reference and the empty pan. The scans were run from 310 K to 330 K at a 10 degree/minute heatup rate and the peak amplitude determinations were made at 320 K. In calculating the specific heat of the material, average peak amplitudes were used; i.e., the peak heights were averaged and the mean value used in the following relationship:

$$Cp = \left\{\frac{A_{sample} + A_{pan}}{A_{reference} + A_{pan}}\right\} \left\{\frac{Wgt_{ref.}}{Wgt_{sample}}\right\} \left\{\frac{0.19574\ cal}{gm\ K}\right\} \qquad (7)$$

where,

0.19574 = Cp of sapphire at 320 K

Density measurements were made using a pycnometer. The technique involves weighing the dry sample, weighing the pycnometer full of water, and then weighing the pycnometer containing the sample and water. The volume of the sample is determined by dividing the mass of the water displaced by the sample by the density of the water. The density of the sample equals the mass of the dry sample divided by the volume. The eight samples used for the density measurements were the same as those used in the burnout tests. Nominal specimen measurements were 1/2" x 3/4" x 1/4" thick and they weighed between 3.2 grams and 3.5 grams. Mean values together with the standard deviations were calculated from the measured results for the thermal diffusivity; specific heat and density, and a conductivity value was obtained from these results. The experimental uncertainty in this value was found from Equation 5 and an uncertainty analysis based upon Equation 8. These results are listed in Table 1.

$$W_K = \left\{ \left[\frac{\partial K}{\partial \alpha} W_\alpha \right]^2 + \left[\frac{\partial K}{\partial \rho} W_\rho \right]^2 + \left[\frac{\partial K}{\partial Cp} W_{Cp} \right]^2 \right\}^{0.5} \quad (8)$$

where,

W_K = experimental uncertainty of conductivity
W_α = experimental uncertainty of diffusivity
$W\rho$ = experimental uncertainty of density
W_{Cp} = experimental uncertainty of specific heat

Table 1. THERMAL PROPERTIES FOR PULTRUDED S-GLASS/EPOXY

	Units	Average	Standard Deviation	Experimental Uncertainty
α	(cm^2/sec)	3.70E-03	4.97 E-04	
ρ	(gm/cm^3)	1.950	0.029	
C_p	(cal/gm K)	0.2167	0.0025	
K	(ca./cm K)	1.56E-03		2.12 E-04

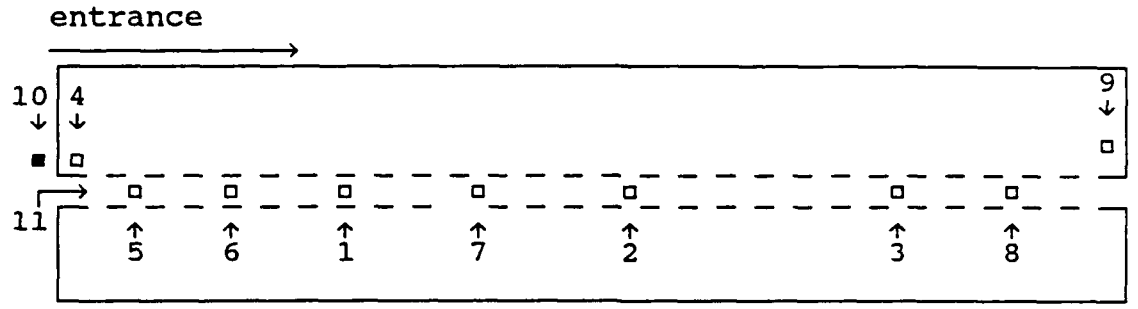
Boundary Conditions

In order to obtain the best possible information from the models, the initial and boundary conditions over which they are solved must be as accurate as possible. Toward this end the boundary conditions were determined experimentally.

An assumption made with respect to the initial condition used in the solutions is that the temperature of the fiber resin/mass as it enters the die is uniform across its thickness. While experiments have shown that the assumption of temperature uniformity through a relatively thin cross section, such as that produced in this study, is reasonable. They have also shown that this temperature will vary depending upon line speed and die temperature. The initial temperature, To, was measured by a stationary thermocouple probe. This probe recorded temperatures on the surface of the composite stock just prior to its entrance into the die and remained in contact with the material throughout the process.

The temperature boundary condition at the die wall was measured by two methods. The first recorded temperatures from in situ surface thermocouples, while the second recorded temperatures from thermocouples imbedded in the die itself. The former method assumes that the surface of the composite is at the same temperature as the die wall. It was a part of the experimental procedure to obtain thermal histories by placing thermocouples both internally and on the surface of the composite as it traveled through the die. In cases where the surface thermocouple maintained its position on the surface of the composite (as determined by post processing examination), the thermal history could be used to define the boundary condition.

The second method required the assumption that the surface of the composite and points within the die very near the surface of the composite are at the same temperature. This assumption is justified since the die is constructed from steel, a much better conductor than the glass/epoxy, and because the thermocouples were so close to the die wall. The holes were drilled in the upper half of the die to a depth approximately 1/16th of an inch away from the die wall. One hole was drilled at the approximate location of the surface exotherm which was calculated from data acquired during previous runs. Three of these holes were used to accommodate die temperature control thermocouples, one for each zone. A linear profile between the sensors was assumed and was incorporated into the surface boundary condition. The positions of these thermocouples are illustrated and described in Figure 1.



Thermocouple	Position
1	die temperature control zone 1, 6.1" from the entrance
2	die temperature control zone 2, 15.8" from the entrance
3	die temperature control zone 3, 23.9" from the entrance
4	entrance die well
5	1.9" from entrance end of die
6	4" from entrance end of die
7	9.6" from the entrance end of the die, location of the surface exotherm
8	27.9" from entrance end of die
9	exit die well
10	die entrance on fiber/resin bundle
11	1.3" inside entrance on die wall surface

Note: The die wells have a 0.355" diameter, are 1.856" deep, are 0.400" from either end, and 0.550" from the sixes of the die (as measured to the circumference).

Figure 1. Thermocouple locations along the length of the die.

Chemical Kinetics of the Epoxy Resin System

As the resin-saturated fiber bundle passes through the heated die, it absorbs thermal energy at a rate which is governed by the thermal characteristics of the composite. Eventually, enough thermal energy is absorbed to initiate the chemical reaction which leads to cross-linking, which in turn results in a cured composite. The contribution of the exothermic reaction to the thermal history of the pultruded composite is represented in the model through the use of a heat source generation term.

The internal heat generated by the curing composite is described by a first order reaction with an Arrhenius temperature dependence. The details of the heat source term are presented in the Appendix. In order to calculate the heat source, certain kinetic parameters are needed including rate of reaction, heat of reaction, and activation energy. The kinetic parameters for the epoxy system used in this study were determined by Zukas.[9] The determination was made using DSC in which he employed both dynamic and isothermal techniques to obtain rate of reaction and extent of reaction information from which the other kinetic parameters are derived.

Zukas reported that when dynamic scanning was used the total heat of reaction decreases as the scan rate is increased. This change was attributed to either the onset of thermal decomposition or a change in the reaction mechanism of the anhydride cured system at higher temperatures. Additional work by Zukas[10] has shown that postcure of pultruded specimens leads to an increase in Tg signifying that thermal degradation was unlikely. In the case of the isothermal experiments, a more constant heat of reaction was observed; however, a decrease in the activation energy as a function of increasing degree of cure was noted. The variation in these values may be due to inadequacies of the Arrhenius expression (see Equation 9) in describing the reaction kinetics of epoxies.

$$\frac{d\alpha}{dt} = (1-\alpha)^n A \exp(-E/RT) \tag{9}$$

Completion of the analysis revealed that a value of 1 for the order of reaction, n, is most appropriate. The results of the analysis are shown in Table 2.

Table 2. KINETIC PARAMETERS FOR THE ANHYDIRDE CURED EPOXY RESIN SYSTEM[9]

Degree Cure (α)	Act. Eng. (E) (kcal/mole)	A at 433 K (1/min)
0.10	19.82	4.73 E+09
0.20	19.45	3.18 E+09
0.30	19.02	2.00 E+09
0.40	18.67	1.37 E+09
0.50	18.34	9.59 E+08
0.60	17.99	6.46 E+08
0.70	17.50	3.75 E+08
0.80	16.78	1.59 E+08
0.90	14.85	1.50 E+07

9. ZUKAS, W. X., and TESSIER, N. J. *Thermal Analysis for Pultrusion Process Modelling.* 32nd International SAMPE Symposium, v. 32, CA, 1987.
10. ZUKAS, W. X. *Monitoring the Cure of an Epoxy-Anhydride Resin.* Polymer Engineering and Science, v. 29, 1989, p. 1153.

If one examines this data in the light of Equation 9, it can be assumed that the impact of the activation energy is proportional to $(1-\alpha)$. With this in mind, a weighted average of the form,

$$E = \left\{ \sum_{i=1}^{n} (1-\alpha_i)\ E_i \right\} / \sum_{i=1}^{n} (1-\alpha_i), \qquad (10)$$

was taken for both the activation energy and the rate constant. The weighted average represents a trade-off between the possibly increased accuracy one might obtain through the use of fitted data to the increase in model performance times gained through the utilization of a constant value for the activation energy and reaction rate. With this in mind, the value for the activation energy used in the model for the epoxy system studied here was 18.67 kilocalories per mole. The same technique of weighted averages was also used for the pre-exponential and this resulted in a value of 2.201 E+09 min^{-1}.

The values obtained from these methods of kinetic parameter determination are for neat resin formulations and may not apply in a strict sense to composite systems. The difference arises from the moderating effect that the reinforcement materials have on the curing kinetics of the resin, as well as possible effects stemming from fiber sizing agents and internal lubricants. It is a common notion in the field of composites that the presence of fillers or fiber lowers the maximum temperature of the exotherm.[11-13] Their effect on the curing reaction is due in part to the mass of the fiber and possibly to surface chemistry. The approach to account for the mass effect is based upon the fiber having replaced resin which would otherwise produce heat during cure. Other effects can be traced to the thermal conductivity of the fiber which is usually much greater than that of the resin. In the materials studied here, for example, the difference was an order of magnitude. The product of the density and the heat capacity is slightly larger for the fiber indicating it would absorb more heat than the resin. Another consideration stems from the fact that all the fibers are aligned in one direction so that a higher degree of order in the nesting of fibers and direction of heat conduction occurs, further minimizing resin mass effects.

The kinetic parameters of the composite system used in this study were estimated from the proportion of neat resin in the composite. Accordingly, a general form of the rule of mixtures involving volume fractions was followed to calculate the rate of reaction, as well as attempting to account for surface and thermal effects of fiber and filler. In the case of the heat of reaction, weight fractions were used to account for the mass effect.

Model Accuracy and Run Times

The accuracy of the models is demonstrated in Figure 2 which compares the experimentally determined thermal history of the pultruded stock at its center to the thermal history as predicted by the lumped parameter, one-dimensional, and two-dimensional models. The case examined here involves a line speed of 6" per minute and a die control temperature of 330°F.

11. MIJOVIC, J. *Cure Kinetics of Neat Versus Reinforced Epoxies.* Journal of Applied Polymer Science, v. 31, 1986.
12. DUTTA, A., and RYAN, M. E. *Effect of Fillers on Kinetics of Epoxy Cure.* Journal of Applied Polymer Science, v. 24, 1979.
13. NG, H., and MANAS-ZLOCZOWER, I. *Kinetic Studies of a Composite Thermoset Cure Reaction - Application in Pultrusion Simulations.* Polymer Engineering and Science, v. 29, no. 5, 1989.

Since the results of the models overlap each other, it is apparent, at least in this particular case, that the dimensionality of the model is not important. The lumped parameter model behaves unrealistically during the initial, very steep portion of the curve. This behavior is short-lived and in all probability is an artifact of the assumed parabolic profile used in solving for the lumped temperature.

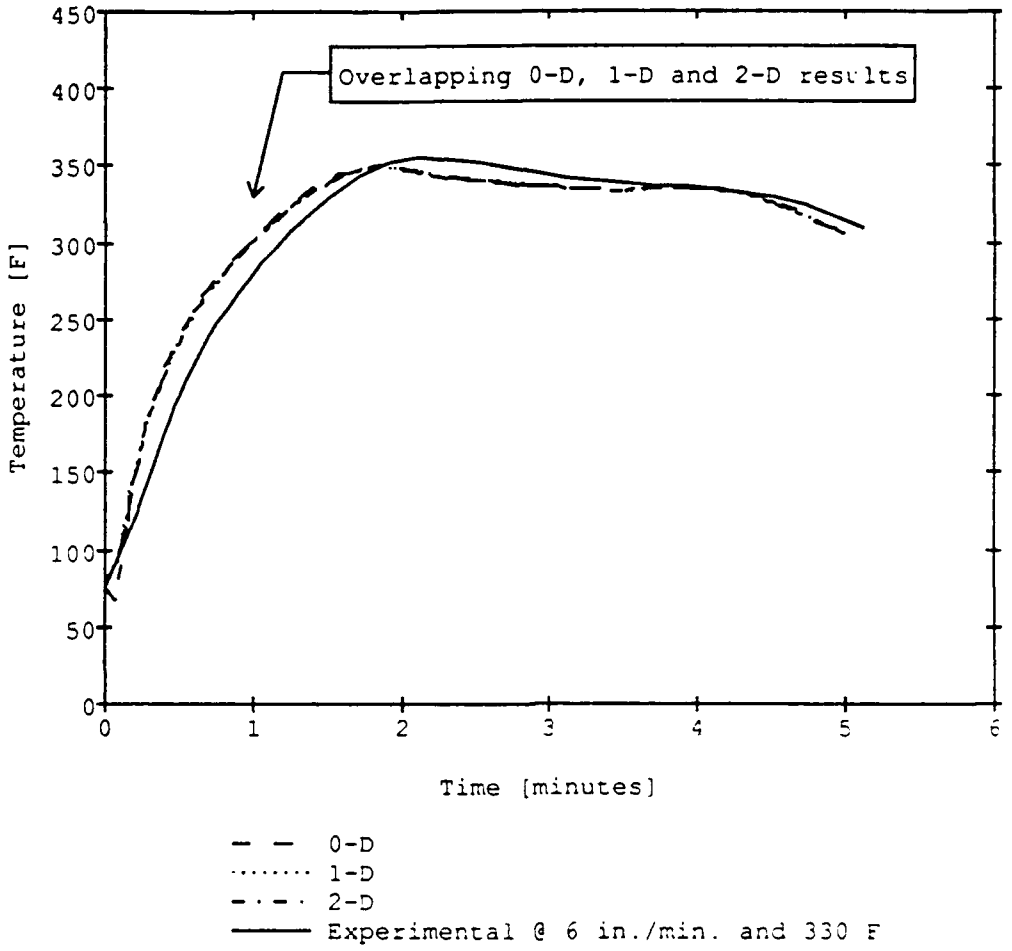


Figure 2. Comparison of results from the lumped parameter, one-dimensional, and two-dimensional models with experimental results for stock pultruded at 6 in./min with die temperature controls set at 330°F.

The results predict the peak exotherm temperature to within about 5°F although the prediction of when/where this maximum occurs was premature. It is somewhat disturbing that even before the exotherm becomes a factor, the predicted temperatures and actual temperatures are not closer together. This area should be governed by heat transfer and not chemical kinetics. Other models such as that developed by Batch and Macosko[14] use a first order time lag factor to account for similar discrepancies which they felt might be due to neglecting axial heat flow, especially where heating rates were high. It is also possible that discrepancies

14. BATCH, G. L., and MACOSKO, C. W. *Heat Transfer and Cure Analysis for Pultrusion*. Presented at SME Pultrusion Clinic, Los Angeles, CA, April 1989.

were due to differences between the measured thermal properties which were obtained for a fully cured composite at room temperature and actual thermal properties at lower degrees of cure and processing temperatures. Mijovic[15] has shown that these differences can be quite large for epoxy resins.

In this case, it seems that using values for heat capacity and thermal conductivity which correspond more closely to conditions of interest occurring early in the process would better serve to improve model results than employing a lag factor. Figures 3 through 7 are comparisons of estimations which were made using the one-dimensional model together with experimentally determined temperature profiles. They constitute a variety of temperature profiles and line speeds and all are based upon the center line temperature of the composite. The material properties used were experimentally determined as were the weight and volume fractions. It can be seen from these figures that the model's prediction of the exotherm temperature is consistently within the ± 10°F range. However, there is a lack of consistency, as illustrated by the fact that in Figure 4 the model underestimates the experimental peak temperature. In Figure 6 the prediction is exact, and in the others (see Figures 3, 5, and 7) the peak temperature is overestimated. A plausible explanation lies in the fact that the kinetic parameters were determined in isothermal experiments carried out at 340°F, which do not account for possible changes in the reaction mechanism as a function of cure temperature. The ability of the model to predict the position of the peak temperature follows a similar course, but maintains accuracy to within about eight seconds. These effects emphasize the need for tempering the interpretation of the model results with experience.

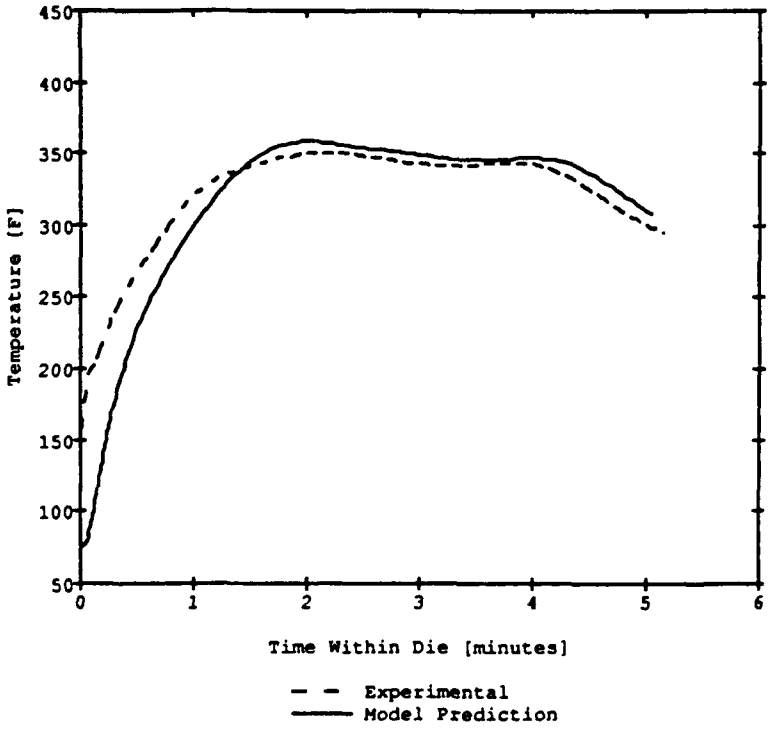


Figure 3. Comparison of model and experimental results for stock pultruded at 6 in./min and a die temperature control of 340°F.

15. MIJOVIC, J., and WANG, H. T. *Modelling of Processing of Composites Part II - Temperature Distribution During Cure.* SAMPE Journal, v. 24, no. 2, 1988.

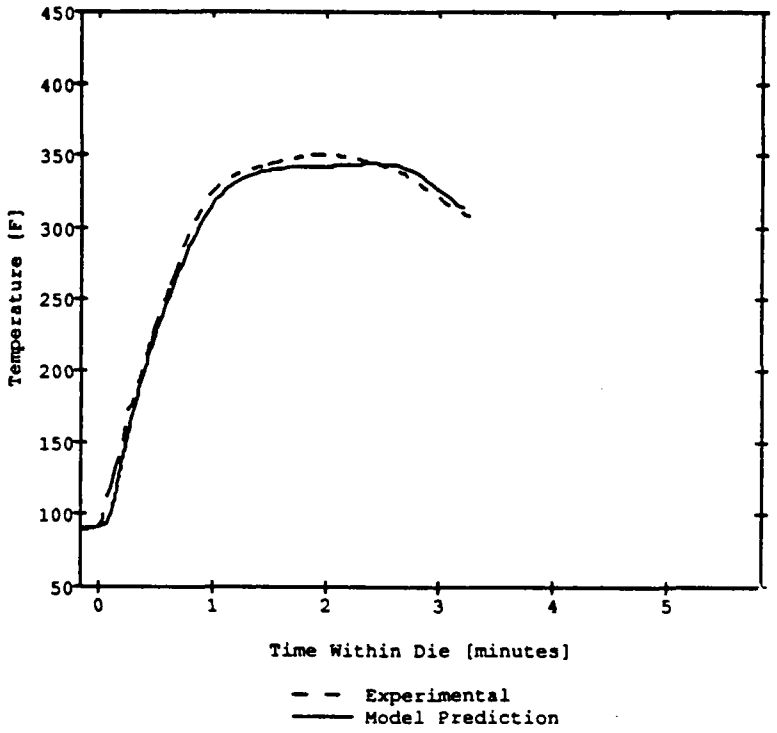


Figure 4. Comparison of model and experimental results for stock pultruded at 9 in./min and a die temperature control of 330°F.

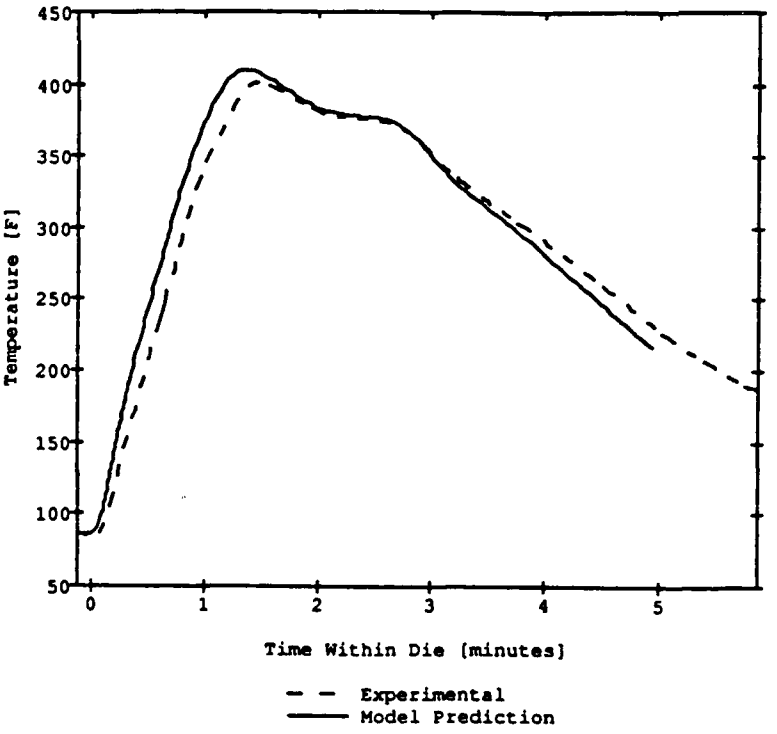


Figure 5. Comparison of model and experimental results for stock putruded at 9 in./min and a die temperature control of 370°F.

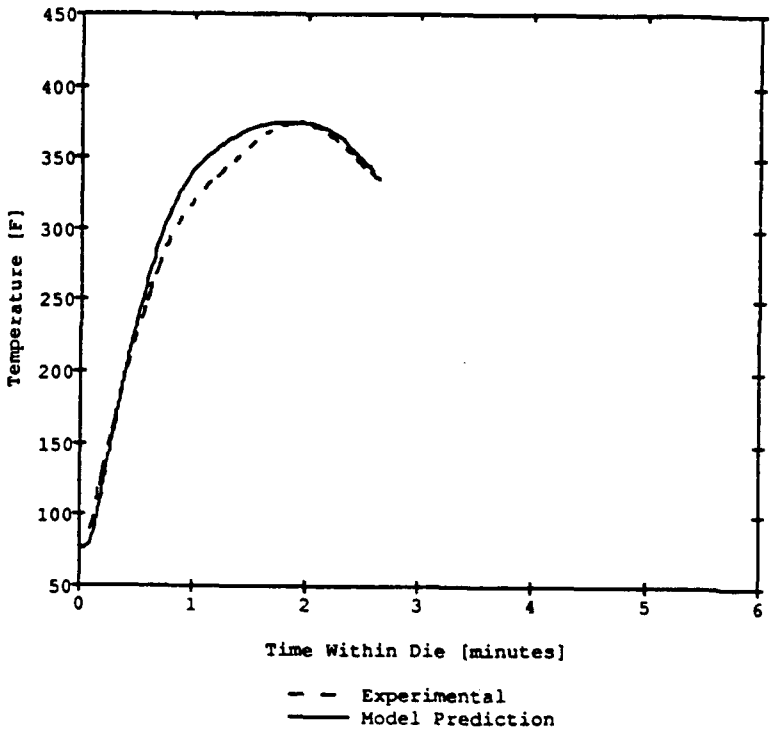


Figure 6. Comparison of model and experimental results for stock pultruded at 12 in./min and a die temperature control of 350°F.

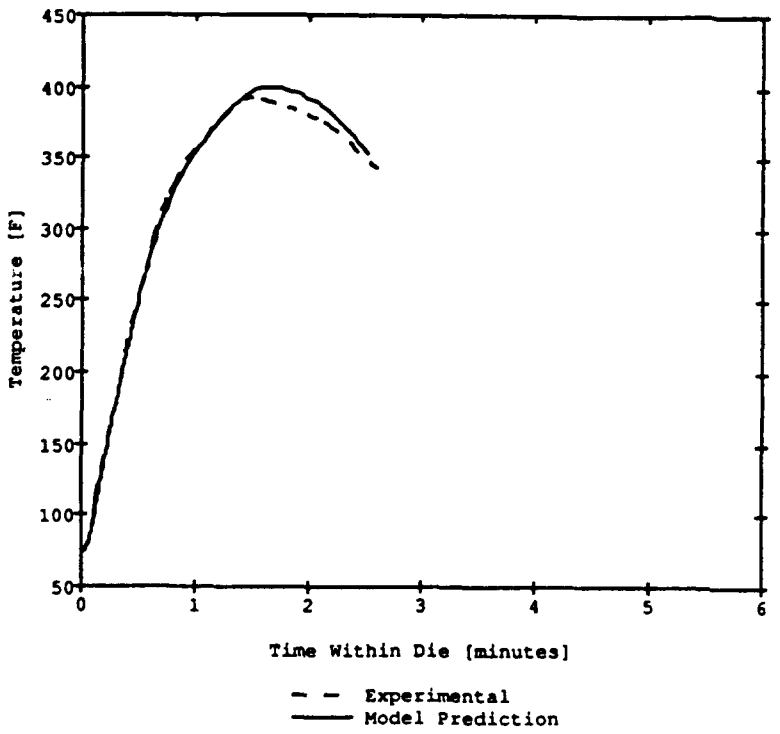


Figure 7. Comparison of model and experimental results for stock putruded at 12 in./min and a die temperature control of 370°F.

Since it is proposed that surface temperatures be measured by means other than embedded thermocouples, the model has been run with die wall boundary conditions generated by both an embedded thermocouple and by thermocouples situated in the die. Figure 8 shows that the results for both cases are very similar, and that the surface temperature can be obtained easily through the use of thermocouples situated within the die with only slightly diminished model accuracy.

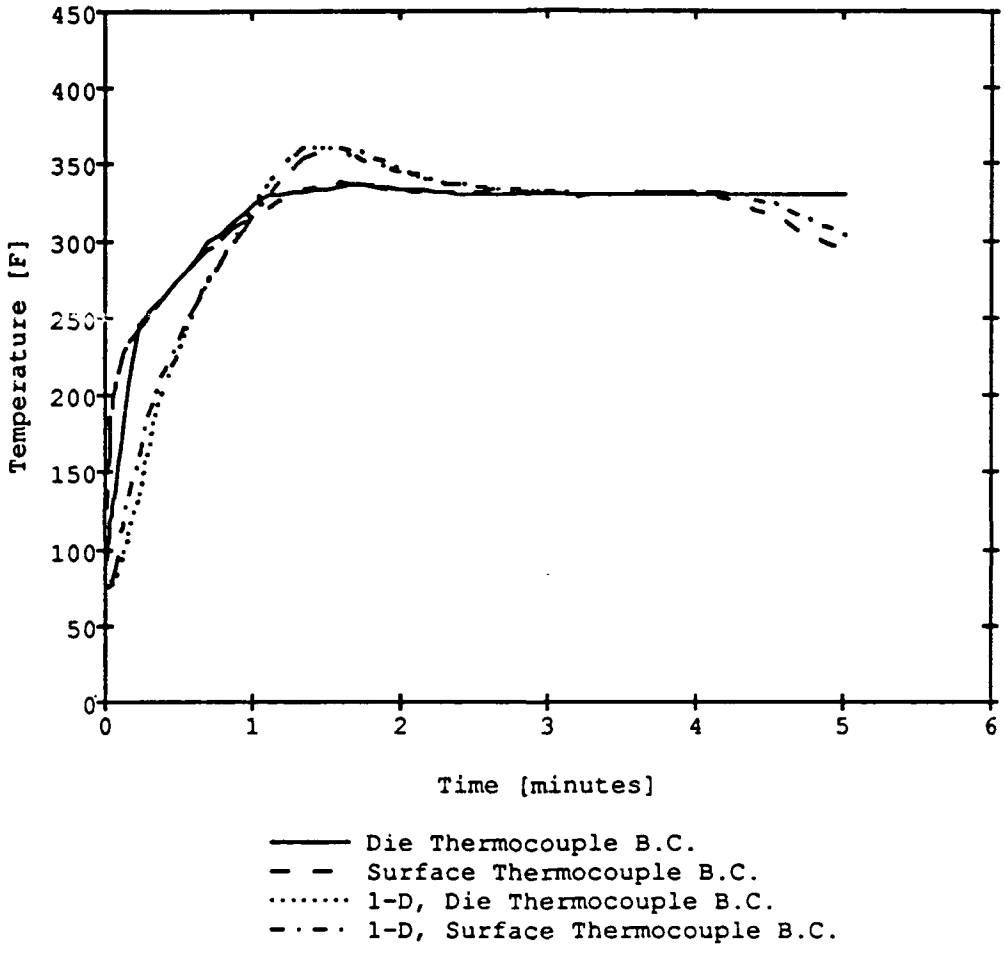


Figure 8. Comparison of results for differing surface boundary conditions.

Another important consideration for the use of the models in a real-time scenario is the actual run time needed to produce the thermal results. The models were run on two different computers; an 80286 based AT class machine with a clock speed of 6 MHz, and an 80386SX machine with a clock speed of 16 MHz. The 386SX computer is a hybrid which uses an 80386 CPU but relies on the 16 bit AT bus structure. Both machines were equipped with the appropriate math coprocessor.

The time required to execute the various models was recorded and the results are presented in Table 3. The lumped and one-dimensional model were run with 700 time steps, and the two-dimensional model was run with 300 time steps. The grid in the thickness or x direction for the one-dimensional model consisted to 40 nodes. The grid in the thickness direction for the two-dimensional model consisted of 11 nodes, while the grid in the width or y direction consisted of 41 nodes. Note that the times shown in Table 3 are only approximate execution times since different brands of a particular computer configuration may have different operating efficiencies. There are possibilities for making considerable improvements in execution time which do not require improved hardware. One such possibility would be the strategic use of assembler subroutines. It is also possible that considerable improvements in execution time could be made through the use of automatically optimized time steps and node distributions.

Table 3. MODEL RUN TIMES

	6 MHz (80286)	16 MHz (80386SX)
Lumped Parameter	8.4 (sec)	2.8 (sec)
One-Dimensional	91.5 (sec)	28.7 (sec)
Two-Dimensional	162.0 (min)	42.3 (min)

A sensitivity study was carried out producing the results illustrated in Figures 9 and 10. These figures were generated by running the one-dimensional model using various combinations of the experimentally determined set of values for thermal and kinetic properties multiplied by a factor. The case in which a factor of 1.0 is applied to all values will be referred to as the standard case (the case in which experimentally determined values were used in the model). In all other cases summarized in Figures 9 and 10, a factor of 1.0 was applied to all values with one exception. This single value was varied through 80%, 90%, 95%, 100%, 105%, 110%, and 120% of its experimentally determined value. In this study, the amount of variation in the properties examined was chosen to be representative of possible experimental error inherent in the determination of values for thermal properties and kinetic parameters. Fortunately, accurate determinations of these properties from the final composite is relatively easy.

Figure 9 graphs the difference in position at which the maximum centerline composite temperature is attained relative to the position of maximum temperature for the standard case. It is plotted as a function of the percentage by which the relevant property was varied. From the graph it is clear that rather large changes in either the density or the thermal conductivity have very little consequence on the model's prediction of the position of the peak temperature. Changes in the heat of reaction, rate of reaction, and heat capacity, however, have larger effects, all of similar magnitude. These effects, although larger than those resulting from changes in density or thermal conductivity, are still small and only on the order of a few seconds. As the value for the heat capacity is increased the position of the composite's maximum centerline temperature is moved downstream toward the die exit. In the case of the rate of reaction and the heat of reaction, the inverse is true.

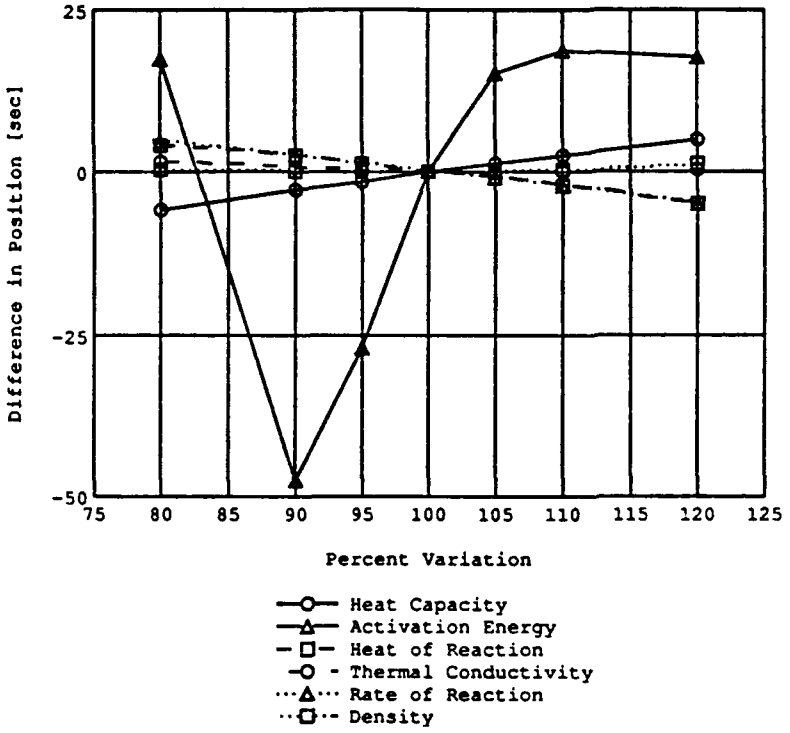


Figure 9. Sensitivity study with respect to position of maximum temperature.

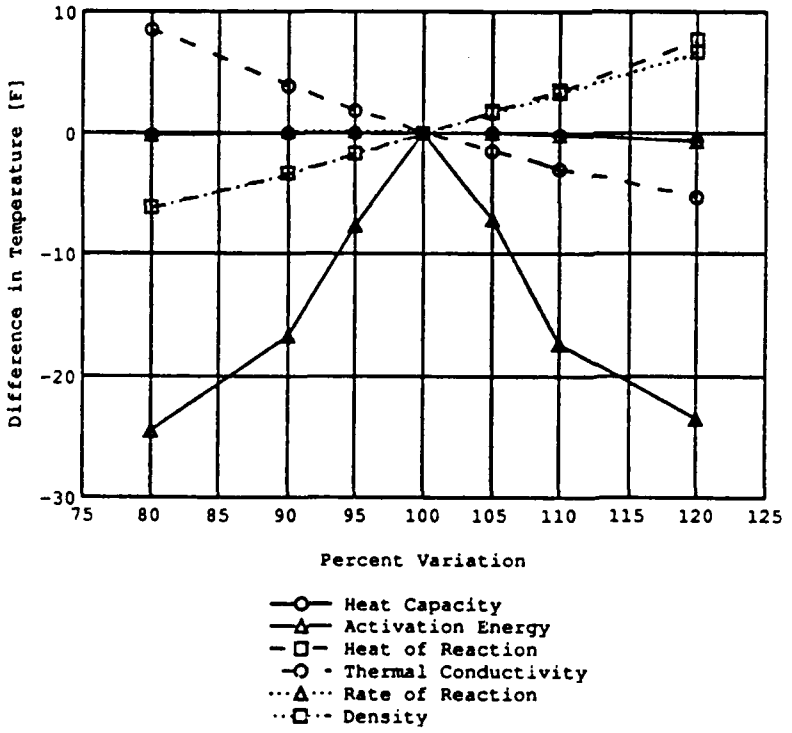


Figure 10. Sensitivity study with respect to maximum centerline temperature.

The parameter with the most impact on exotherm position in the 80% to 120% range is activation energy. The data point graphed for the 80% value of the activation energy appears strange since it is unexpected that the position of the exotherm would suddenly be moved further along the die when the activation energy is decreased. This is correct, however, and is due to the fact that in all other cases the maximum temperature is associated with the exotherm. In the 80% case, however, the model predicts the exotherm occurring while the composite is still fairly cool, and the added heat is not enough to cause the composite temperature at the point of exotherm to exceed the ultimate composite temperature. Therefore, the position of the maximum temperature in the 80% case is not associated with the exotherm but rather with the composite approaching the die temperature. Figure 11 is a graphical representation of this situation, which also shows that as the activation energy increases the limiting case is approached in which there is no exotherm.

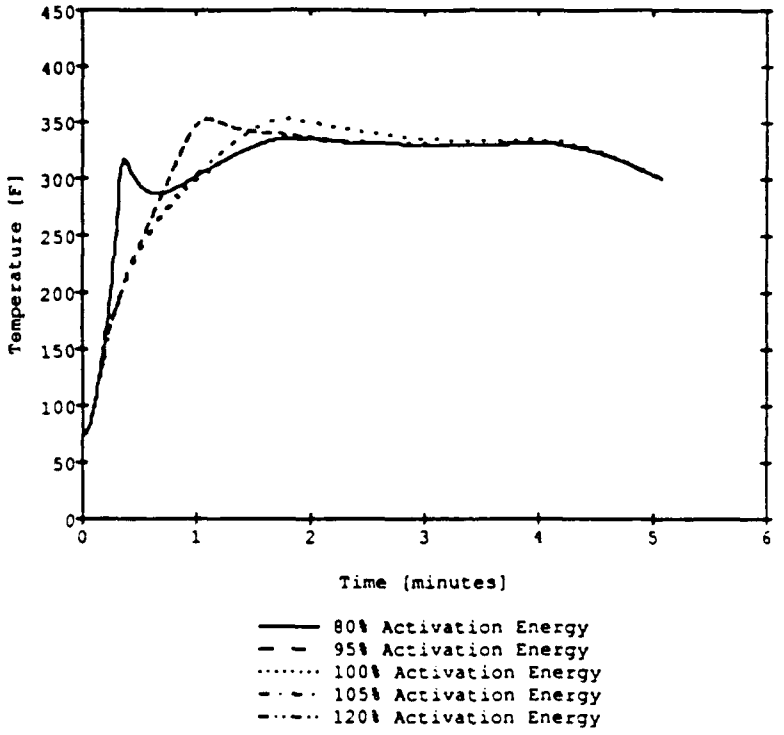


Figure 11. Effect of variation of activation energy.

Figure 10 is analogous to Figure 9 and plots differences in the maximum temperature achieved at the center of the composite relative to the standard case. Changes in the rate of reaction and the heat capacity have almost no effect on the maximum temperature. In contrast, changes in conductivity, heat of reaction, and density produce greater changes in maximum temperature. Increases in the thermal conductivity lower the maximum temperature while increases and decreases in the heat of reaction or density result in greater maximum temperatures.

Once again, one parameter dominates; i.e., the activation energy. A 10% change in this term produces almost a 20°F change in the maximum centerline temperature. This is readily explained through an examination of Equations 1 and 2 which serve as the basis of the model. This effect is anticipated since the activation energy is a term in an exponential function.

It is important to note that these results have been arrived at through the use of an experimentally obtained die boundary condition which is coupled to the properties of the pultruded material. The deviations observed are not predictors of the effect variations of properties will have on the process. They should be viewed as indicators of the effects that errors in the specification of thermal and kinetic property values will have upon the results of the model. As such, they allude to the difficulty or ease in obtaining property data which is of the required accuracy.

Implementation of the Process Model

There are limitations inherent in this model, as in all models describing complex processes, since it is virtually impossible to completely describe a complex process mathematically. On the other hand, this effort has also shown that it is possible to obtain rather accurate results in rapid time on a PC based computer platform. The simplest manner in which this process model might be implemented is in a predictive capacity. This scenario involves running the model prior to actually operating the pultruder in order to obtain a reasonable and operable group of first iteration set points. In this sense, the model would allow one to inexpensively and quickly simulate the early trial and error experiments that have come to be associated with composite processing.

The technique for using the system in a predictive manner involves obtaining or developing the kinetic parameters, material properties, and the boundary conditions which, in this case, is the die temperature profile. The thermal properties are often available from material manufacturers or material handbooks, and as shown in the previous section, the solution is forgiving of moderate variations in their values. Good approximations of thermal properties can be obtained through such methods as the rule of mixtures or the Halpin-Tsai equations.[16] The same cannot be said for kinetic data. This information can be obtained, though, from relatively inexpensive equipment such as a DSC. Once the parameters for the model have been determined variations in process conditions or fiber/resin ratios can be accommodated, in a sense amortizing the effort to acquire the parameters.

The final requirement for making estimations with the model is a set of boundary conditions. The model results offered in this report were based upon a die temperature profile which was acquired during actual processing. This data is not available for the initial run and so must be estimated. This will require some operator experience in that the manner in which a die heats up is dependent upon its mass and geometry. It should be realized, however, that this first model run is designed to serve as an initial iteration in a series of runs which will be used to determine the final set point.

16. HALPIN, J. C., and TSAI, S. W. *Environmental Factors in Composite Materials Design*. U.S. Air Force Materials Laboratory Technical Report No. AFML-TR-67-4223, 1967.

As an illustration of such a technique the density, heat capacity, and thermal conductivity for the E-glass/epoxy pultrusion were estimated using the rule of mixtures and the Halpin-Tsai equations. A die temperature profile using essentially five data points was estimated by linear interpolation between the five specified points. These points define the temperature distribution in the die as it ramps from its initial temperature to an intermediate temperature, resulting from the rapid increase of heating rate which occurs in the entry portion of the die, to the die control temperature which is maintained for a period of time, to a cooler temperature at the die exit. For this particular example, the line speed chosen was 6 in./min and the estimated die temperature profile was specified for die temperature control set points of 340°F at each zone. This data was then used to run the model, the results of which are illustrated in Figure 12.

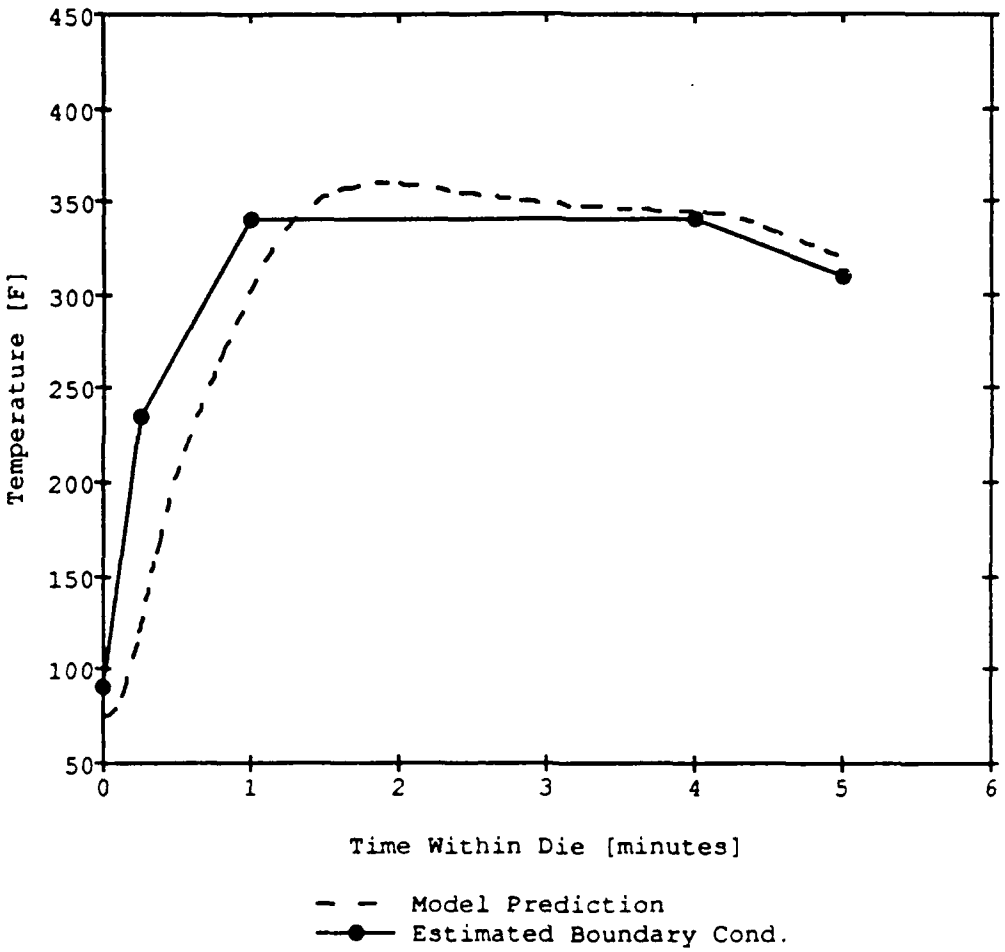


Figure 12. Initial estimation of centerline thermal history for stock pultruded at 6 in./min and a die control temperature of 340°F.

Any number of line speed and die temperature profiles could have been selected for analysis as potential set points. The fact that the model runs so quickly encourages this. Once the curves representing the predicted temperature history of the pultruded stock at the anticipated set points have been generated by the model, the second stage of the process begins.

This involves applying a set of general rules, based upon desirable composite temperature histories, to choose an appropriate set of control temperatures for the die. These general rules may be designed to place limitations on the heat-up rate and characteristics of the exotherm by describing the magnitude and position of the peak temperature excursion. Evidence[17,18] has shown that optimum results for this resin system, as well as some others, are obtained when this excursion does not exceed 25°F. The position at which this occurs is also important, and a good rule of thumb for this resin system is that the gel zone should be situated approximately midway in the die. Since this die is 30" long this would place the desired gel zone at about the 15" mark. It is important to note that the optimum location of the gel zone is in part dependent upon the resin system employed and, therefore, will eventually require some sort of experimental verification.

This portion of the analysis requires some engineering familiarity with the process and the model used to represent it. Figure 12 shows that the peak temperature predicted by the model is 20°F above the die temperature and that this occurs at 1.8 minutes into the run. This translates to a peak exotherm position of about 12" from the die entrance. Since epoxy gelation generally occurs after the exotherm maximum, these results indicate that this set of process variables are potentially applicable.

The next step of the process would be to actually run the pultruder at the selected set points. While the machine is running a new set of actual boundary conditions would be generated. Specifically, the actual initial temperature of the composite and the real-time die temperature profile would be recorded. This data would then serve to either verify the original or establish a new group of set points. This can be illustrated by examining the predicted profile of Figure 3 which is based upon a real-time die temperature profile employing the same line speed and profile estimates used to make Figure 12. Using this data the model predicts the same temperature rise but moves its position about 1-2/3" toward the die exit. Of course, in addition to predictions of the composite temperature profile, a real-time experimental temperature profile could be obtained to serve as a reference or baseline. The procedure is illustrated in the flow diagram shown in Figure 13.

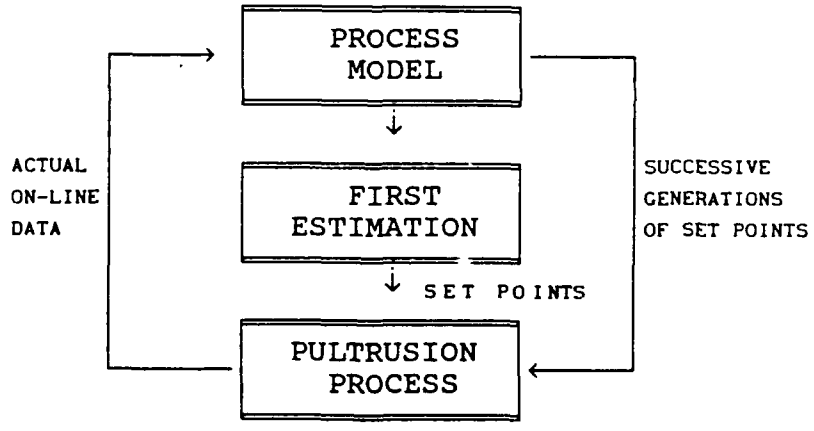


Figure 13. Schematic of set point determination.

17. JAKLITSCH, D. J. *Computer Automation of the Pultrusion Process.* Doctoral Thesis, University of Lowell, Lowell, MA, 1990.
18. OUTWATER, J. O. *On the Mechanics of Pultrusion.* 41st Annual Technical Conference, RP/CI, Society of the Plastics Industry, January 1986.

Proposed Methodology of a Pultrusion Control System

It is apparent that one of the essential components of the system just described is, in the vernacular of computer integrated manufacturing, warmware or human beings. The operator must make decisions as to how and when alterations to the set points should be implemented. As is the case with many systems such decisions are required to prevent system instability. The computer has the ability to make a vast number of decisions in a very short timeframe which ignore transients and the need for settling times in real systems regardless of the consequences. This, however, may be reconciled in the control software.

The logical evolution of a system of this nature would be to invoke a decision maker, as in a rule based expert system. A working decision maker would either serve to take over some of the tasks now performed by the operator or provide a tool to assist the operator in the decision making.[19] Such a system would be founded on an empirical data base based upon scientific principle or experience. It might include such general rules as the 25°F rule which was used to test the peak exotherm, limits as to where in the die this peak should occur, and how rapidly changes could be invoked. Other inclusions would be concerned with limitations imposed by the pultrusion hardware, such as die heating and cooling rates, and alarm states indicating failures in the die heaters or problems associated with the line speed.

Although in situ sensor information which directly relates pultruded properties to control parameters is currently not practical, there are possibilities for obtaining real-time process information using a variety of other sensors. Fiber tension in the creel and resin bath viscosity, although of minimal importance to the resin system examined here,[20] may be important in other fiber/resin systems and therefore worth monitoring. This information, as well as thermal data, offered by thermocouples imbedded in the die, and measurements of pulling force could serve as input for the control system. Recently, Parker,[21] at NASA Langley, developed ultrasonic sensors which can be placed in the die in order to obtain ultrasonic B-scan data in real-time. The fact that these measurements are related to the viscosity and degree of cure of the resin demonstrates the potential of such information in a control system. The rule based system would exist as a computer program which interprets computer inputs as well as the model's output and could be described schematically, as illustrated in Figure 14.

An intrinsic aspect of such a system would be its continuing evolution. As more information was obtained, as new phenomena were identified, and as sensors or machine components were improved, the system would grow and hopefully improve. There are limitations as to what can be included in an expert system since the rules must be equations or experiences that can be expressed in a form which the software can interpret and use. This does not preclude the addition of rules which are somewhat ill-defined or uncertain. An instrument for handling such situations exists in the form of fuzzy logic which allows one to express qualitative terms in a mathematical manner. The method allows one to represent real-world events as a continuous spectrum of values by using fuzzy sets. Unlike boolean sets, however, where elements are either in the set or not, that is true or false, fuzzy logic elements can have partial

19. SERVAIS, R. A., LEE. C. W., and BROWNING, C. E. *Intelligent Processing of Composite Materials.* SAMPE Journal, v. 22, no. 5, 1986.
20. JAKLITSCH, D. J. *Computer Control of the Pultrusion Process.* Doctoral Dissertation, University of Lowell, Lowell, MA, 1990.
21. PARKER, F. R. *In Process Ultrasonic B-Scan of a Pultruding Graphite/Epoxy Composite.* Proceedings of the Fiber-Tex 1990 Conference, Clemson, SC, August 1990.

membership. This technique has also been proposed for use in a knowledge-based autoclave control system.[22] A consequence of using more than one of these techniques might be a very large set of rules. These rule bases when queried may suggest more than one course. There must be some mechanism for choosing which course or, if they are compatible, courses of action should be taken. A separate program would have to be formulated in order to perform arbitration and avoid conflicts. This necessitates a more complicated decision tree, the function of which would be to assimilate all input and produce a control system. It would integrate the process model, knowledge base, and sensor input with the actual process.

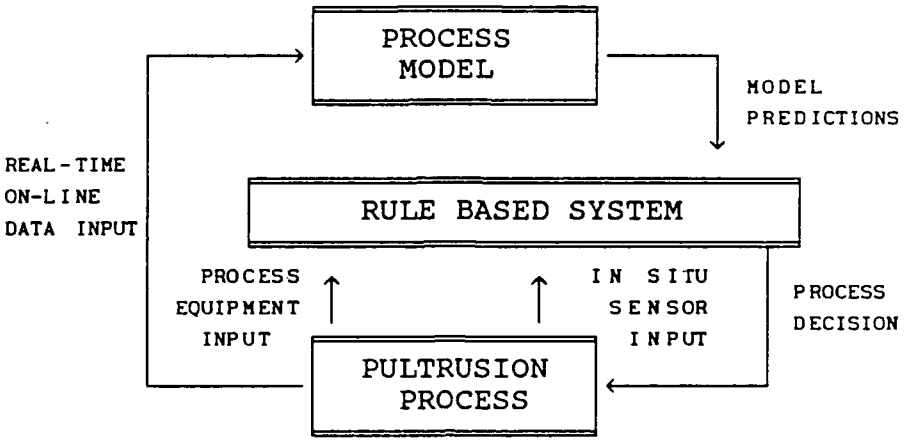


Figure 14. Incorporation of components in proposed decision making pultrusion control system.

A foreseeable system would integrate a mathematical model coupled with experience and, to fill in the gaps, intuition. Implementing this requires the development of a data base consisting of property data and processing hardware characteristics, the definition of a rule base derived from experience, and the establishment of quality control criteria. These elements in conjunction with a real-time data acquisition module would be managed by a decision tree ultimately determining process setpoints.

The schematic shown in Figure 15 represents an outline of such a system. The speed and true multitasking ability available with state of the art PC based computer platforms is opening the door for such techniques. There are also hardware and software products currently on the market which allow one to develop fuzzy logic control systems on the PC platform.

22. WU, H. T., and JOSEPH, B. *Knowledge Based Control of Autoclave Curing of Composites.* SAMPE Journal, v. 26, no. 6, 1990.

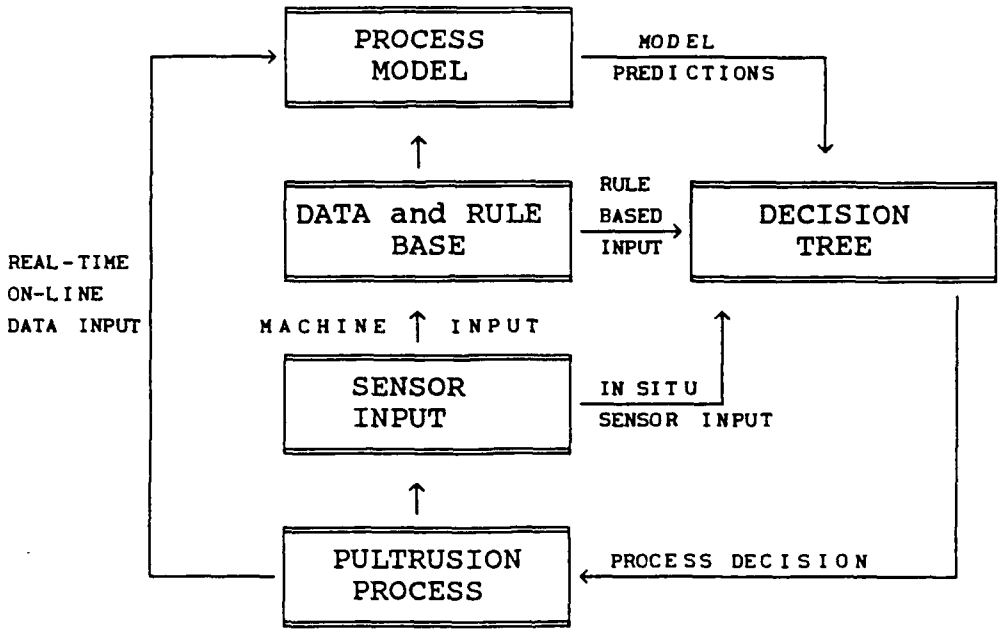


Figure 15. Proposed pultrusion control system.

CONCLUSIONS

Control of the pultrusion process through the use of a process model does have near-term potential in real-time applications. The model results and conditions under which they were attained meet, or have the potential to meet, the criteria for feasibility as outlined in the introduction. If the material and kinetic properties are not already available in the literature, they can be obtained through fairly simple and quick methods. Since the lumped parameter and one-dimensional and two-dimensional models give practically identical results, any would be appropriate and both the lumped parameter and one-dimensional have execution times in the real-time domain. Insofar as computing machinery expense in concerned, the PC based platform represents the low end for computer cost. The accuracy of the model was somewhat limited, but possibly adequate, and it remains a viable tool through its predictive capacity. Recovery from computer errors has not been addressed. However, this issue is routinely solved in other applications and should also be tractable in this situation.

Future study should be directed towards increasing the accuracy of the model and increasing the processing speed. Efforts should also be extended towards the incorporation of machine and sensor input into a robust control system.

ACKNOWLEDGMENTS

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APPENDIX

The one-dimensional model was developed using the methodology established by Patankar (see Reference 2). It makes use of the control volume formulation in which the calculation domain is divided into a number of nonoverlapping control volumes such that there is one control volume surrounding each grid point. The differential equation is integrated over each control volume by using piecewise profiles to express the variation in the dependent variable between the grid points. The result is the discretization equation describing the relationship between the values of the dependent variable for a group of neighboring grid points. Since the source term is a nonlinear function of the dependent variable, the source term dependence is linearized. This allows the discretization equations to be solved by linear algebraic techniques.

The following discussion outlines the technique for deriving the discretization equation. Figure A1 illustrates the grid point cluster used to solve the one-dimensional problem.

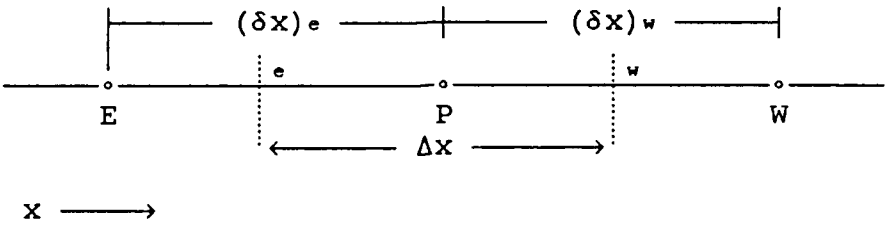

Figure A1. The grid point cluster used for the model solution.

The energy equation:

$$\rho Cp \int_e^w \int_t^{t+\Delta t} \frac{\partial T}{\partial t}\, dt\, dx = \int_t^{t+\Delta t} \int_e^w k \frac{\partial}{\partial x}\left(\frac{\partial T}{\partial x}\right) dx\, dt + \int_t^{t=\Delta t} \int_e^w S\, dx\, dt \qquad \text{(A1)}$$

$$\Rightarrow \rho Cp \int_e^w \left(T(x) - T_t(x)\right)_{t+\Delta t} dx = \int_t^{t+\Delta t} \int_e^w k \frac{\partial}{\partial x}\left(\frac{\partial T}{\partial x}\right) dx\, dt + \int_t^{t+\Delta t} \int_e^w S\, dx\, dt \qquad \text{(A2)}$$

Let,

$$T@\ t+\Delta t = T'$$

$$T@\ t = T^{\circ}$$

$$\Rightarrow \rho Cp \int_e^w \left\{(T'(x) - T^{\circ}(x)\right\} dx = \int_t^{t+\Delta t} \int_e^w k \frac{\partial}{\partial x}\left(\frac{\partial T}{\partial x}\right) dx\, dt + \int_t^{t+\Delta t} \int_e^w S\, dx\, dt \qquad \text{(A3)}$$

Since T(x) is constant over the interval from e to w and, furthermore, is taken to be equal to that at node P, we say that

$T(x) = T_P = \text{constant.}$

$$\Rightarrow \rho Cp \left(\int_e^w T'(x)dx - \int_e^w T^\circ(x)\,dx \right) = \int_t^{t+\Delta t} \int_e^w k \frac{\partial}{\partial x}\left(\frac{\partial T}{\partial x}\right) dx\,dt + \int_t^{t+\Delta t} \int_e^w S\,dx\,dt \tag{A4}$$

$$\Rightarrow \rho Cp\,(\Delta x\,T'_P - \Delta x\,T^\circ_P) = \int_t^{t+\Delta t} \int_e^w k \frac{\partial}{\partial x}\left(\frac{\partial T}{\partial x}\right) dx\,dt + \int_t^{t+\Delta t} \int_e^w S\,dx\,dt \tag{A5}$$

$$\Rightarrow \rho Cp\,\Delta x\,(T'_P - T^\circ_P) = \int_t^{t+\Delta t} \left(k_w \frac{\partial T}{\partial x}\Big|_w - k_e \frac{\partial T}{\partial x}\Big|_e \right) dt + \int_t^{t+\Delta t} \int_e^w S\,dx\,dt \tag{A6}$$

At this point we find an expression for the slope of T at e and at w. It should be noted that E and W stand for the nodes, and that e and w stand for the interfaces between the control volumes. A linear expression was assumed for the slope.

Let,

$$\frac{\partial T}{\partial x}\Big|_e = \frac{T_P - T_E}{\partial x_e}$$

$$\frac{\partial T}{\partial x}\Big|_w = \frac{T_W - T_P}{\partial x_w}$$

$$\Rightarrow \rho Cp\,\Delta x\,(T'_P - T^\circ_P) = \int_t^{t+\Delta t} k_w \left(\frac{T_W - T_P}{\partial x_w}\right) dt - \int_t^{t+\Delta t} k_e \left(\frac{T_P - T_E}{\partial x_e}\right) dt + \int_t^{t+\Delta t} \int_e^w S\,dx\,dt \tag{A7}$$

$$\Rightarrow \rho Cp\,\Delta x\,(T'_P - T^\circ_P) = \int_t^{t+\Delta t} k_w \frac{T_W}{\partial x_w}\,dt - \int_t^{t+\Delta t} k_w \frac{T_P}{\partial x_w}\,dt - \int_t^{t+\Delta t} k_e \frac{T_P}{\partial x_e}\,dt + \int_t^{t+\Delta t} k_e \frac{T_E}{\partial x_e}\,dt + \int_t^{t+\Delta t} \int_e^w S\,dx\,dt \tag{A8}$$

The fully implicit model assumes that the temperature from t to $t+\Delta t$ is equal to that at time $t+\Delta t$, therefore, Tw, Tp, and Te may be treated as constant over the interval $t \rightarrow t+\Delta t$.

$$\rho C p\,\Delta x\,(T'_P - T^\circ_P) = \frac{k_w}{\partial x_w}\Delta t\,(T_W' - T_P') + \frac{k_e}{\partial x_e}\Delta t\,(T_W' - T_P') + \int_t^{t+\Delta t} \int_e^w S\,dx\,dt \tag{A9}$$

To linearize the source term, let $S = S_c + S_P T$

$$\Rightarrow \rho Cp\,\Delta x\,(T_P' - T_P^\circ) = \frac{k_w}{\partial x_w}\Delta t\,(T_W' - T_P') + \frac{k_e}{\partial x_e}\Delta t\,(T_E' - T_P') + \int_t^{t+\Delta t} \int_e^w S_c + S_P \cdot T\,dx\,dt \tag{A10}$$

$$= \frac{kw}{\partial X_w} \Delta t \ (Tw' - TP') + \frac{ke}{\partial Xe} \Delta t \ (TE' - TP')$$

$$+ \int_t^{t+\Delta t} \int_e^w Sc \ dx \ dt + \int_t^{t+\Delta t} \int_e^w SP \cdot T \ dx \ dt$$

$$= \frac{kw}{\partial Xw} \Delta t \ (Tw' - TP') + \frac{ke}{\partial Xe} \Delta t \ (TE' - Tw')$$

$$+ \Delta x \ \Delta t \ Sc + SP \int_t^{t+\Delta t} \int_e^w T \, dx \ dt$$

Recalling that Sc and SP are constants,

$$\rho Cp \ \Delta x \ (TP' - TP^\circ) = \frac{kw}{\partial Xw} \Delta t \ (Tw' - TP') + \frac{ke}{\partial Xe} \Delta t \ (TE' - TP') + \Delta x \ \Delta t \ Sc + \Delta x \ \Delta t \ SP TP' \tag{A11}$$

Dividing through by Δt yields,

$$\rho Cp \frac{\Delta x}{\Delta t} (TP' - TP^\circ) = \frac{kw}{\partial Xw} (Tw' - TP') + \frac{ke}{\partial Xe} (TE' - TP') + \Delta x \, Sc + \Delta x \ SP TP' \tag{A12}$$

This can be rearranged to give

$$(\rho Cp \frac{\Delta x}{\Delta t} + \frac{kw}{\partial Xw} + \frac{ke}{\partial Xe} - SP \ \Delta x) TP' = \frac{kw}{\partial Xw} Tw' + \frac{ke}{\partial Xe} TE' + \Delta x \ Sc + \rho Cp \frac{\Delta x}{\Delta t} TP^\circ \tag{A13}$$

Let,

$$aw = \frac{kw}{\partial Xw}$$

$$ae = \frac{ke}{\partial Xe}$$

$$aP^\circ = \rho Cp \frac{\Delta x}{\Delta t}$$

$$aP = aP^\circ + aw + ae - SP \ \Delta x$$

$$b = aP^\circ + TP^\circ + Sc \ \Delta x$$

Then,

$$aP \ TP' = aw \ Tw' + ae \ TE' + b \tag{A14}$$

It should be noted that if all nodes are equidistant, and all control volume faces are centered between nodes, and if the thermal conductivity is not a function of temperature, then $\Delta x = \partial x_e = \partial x_w$ and $k_e = k_w \Rightarrow a_w = a_e = k/\Delta x$.

One last comment is that the central boundary condition is a special case using a half control volume where node one represents the center of the composite.

In lieu of presenting a printout of the computer code used to solve the above equations, the authors wish to make it available via floppy disk. Anyone wishing a copy of the one-dimensional or two-dimensional models written in Turbo Pascal® should contact the authors.

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Donald J. Jaklitsch, and Maria T. Bostic
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